



Loop growth and point defect profiles during HVEM irradiation

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Abstract In this work the point-defect profile in the thin foil has been included in the model for the growth of dislocation loops during HVEM irradiation suggested by Kiritani, Yoshida, Takata, and Maehara [1], and the possible effect of divacancies is discussed. It is found that there is a fairly wide transition range between the two extreme cases described by Kiritani et al. (the vacancy- and the surface-dominant case); this can directly (without the necessity of a divacancy effect) explain the observation of apparent activation energies for loop growth smaller than $1/2 E_V^M$ (where E_V^M is the vacancy migration energy). Even after the inclusion of the point-defect profiles there are indications that the model cannot fully account for the loop growth behaviour in situations where surface losses and recombination losses are comparable.	Copies to

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1. Introduction

Kiritani, Yoshida, Takata, and Maehara [1] have derived expressions for the growth rate of interstitial dislocation loops during irradiation in a high voltage electron microscope (HVEM). They described two extreme situations: "the vacancy-dominant case" (named so by Barlow and Leffers [2]) with a linear dependence of $\ln \dot{L}$ on $1/T$ (\dot{L} and T being the growth rate and the irradiation temperature, respectively) and the "surface-dominant case" with a temperature-independent growth rate.

It is the aim of the present work to show that there is, between these extreme cases, a fairly wide transition range and that this range includes typical experimental situations. The experimental observations of loop growth in copper also made by Kiritani et al. [1] seem to fall in the transition range. The realization that the experiments of Kiritani et al. are in the transition range brings us to consider certain reinterpretations of their results.

2. Basic Equations for Loop Growth

The work of Kiritani et al. [1] is based on the following equation for the growth of an interstitial dislocation loop:

$$\dot{L} = 2\sigma(Z_{IL}M_I C_I - Z_{VL}M_V C_V) \quad (1)$$

where σ is the change in loop radius by the absorption of one interstitial per atom site along the dislocation, Z_{IL} and Z_{VL} are the absorption cross sections (per atom site) of the loop for interstitials and vacancies, M_I and M_V are the "mobilities" of interstitials and vacancies, and C_I and C_V are the concentrations of interstitials and vacancies (in defects per atom). In equation (1) and in all the equations to follow in sections 2, 3, and 4

the effect of divacancies is neglected. The whole problem of the possible effect of divacancies will be discussed in section 6 on the basis of experimental observations and theoretical considerations.

Kiritani et al. considered the number of point defects absorbed by dislocation loops to be insignificant for the overall point-defect population (i.e. the point-defect concentrations were determined by production rate, diffusion to the surfaces, and recombination). A combination of equation (1) with the equations for the formation and disappearance of interstitials and vacancies leads to the two extreme situations, viz. the vacancy-dominant case and the surface-dominant case for which \dot{L} can be expressed as shown in equation (2) and (3), respectively (equation (3) was not written out explicitly by Kiritani et al.):

$$\dot{L} = 2\sigma(Z_{IL} - Z_{VL})(PM_V/Z_{IV})^{\frac{1}{2}} \quad (2)$$

$$\dot{L} = 2\sigma(Z_{IL} - Z_{VL})P/C_S \quad (3)$$

In these equations P is the production rate of Frenkel pairs, Z_{IV} is the capture-site number for recombination of interstitials and vacancies, and C_S is the surface sink efficiency (assumed constant in a given experiment).

The vacancy-dominant case refers to the situation away from the surface in a sufficiently thick foil. As seen from equation (2) loop growth is a thermally activated process with an apparent activation energy of $1/2 E_V^M$, where E_V^M is the vacancy migration energy (M_V is proportional to $e^{-E_V^M/kT}$, k being Boltzmann's constant).

The surface-dominant case refers to the situation close to the surface. Equation (3) shows that the loop growth rate is independent of temperature for these conditions.

Kiritani et al. determined the apparent activation energy for loop growth in copper to be $0.30\text{eV} \pm 0.02\text{eV}$, which is less than

$1/2 E_V^M$; recent measurements give E_V^M values for Cu of about 0.7eV (Bourassa and Lengeler [3], Wright and Evans [4], and Antesberger, Sonnenberg, and Wienhold [5] found 0.72eV, 0.71eV, and 0.70eV, respectively). Kiritani et al. ascribed this discrepancy to the presence of divacancies with lower migration energy than monovacancies. As it is to be shown, we can, on the basis of the present investigation, explain the results of Kiritani et al. without involving divacancies (for a theoretical discussion of the possible role of divacancies see section 6).

3. Point-Defect Profiles and their Effect on Loop Growth

Foreman [6] has calculated the point-defect concentrations in thin foils during HVEM irradiation as functions of foil thickness and position in the foil. The calculations are based on the assumption that the concentrations are determined by production rate, diffusion to the surface, and recombination, i.e. basically the same assumptions as those in the work by Kiritani et al. [1]. Thus, in order to include the transition range between the two extreme cases treated by Kiritani et al. one would just have to insert in equation (1) the point-defect concentrations calculated by Foreman.

The results of Foreman's calculations [6] were presented as the set of curves reproduced in fig. 1. The curves give the factor by which the equilibrium point-defect concentrations in the interior of an infinitely thick foil should be multiplied to give the actual concentrations. This proportionality factor depends on the distance from the surface divided by the foil thickness (in the following designated d) and the dimensionless parameter f defined by

$$f = Z_{IV} P t^{\frac{1}{2}} / (v_V \gamma_I \gamma_V \lambda^{\frac{1}{2}}) \quad (4)$$

where t is the foil thickness, v_V is the vacancy jump frequency, and γ_I and γ_V are constants relating the interstitial and vacancy diffusion coefficients D_I and D_V and the jump distance λ to the corresponding jump frequencies by the equations

$$D_I = \gamma_I \lambda^2 v_I \quad (5)$$

$$D_V = \gamma_V \lambda^2 v_V \quad (6)$$

It should be noticed that for the parameters which are identical in the calculations of Kiritani et al. and Foreman we have adopted the symbols used by Kiritani et al. In the following we shall use the designation $g(f,d)$ for the Foreman proportionality factor given in fig. 1.

Foreman's calculations (as those of Kiritani et al.) refer to steady-state conditions, i.e.

$$C_I D_I = C_V D_V \quad (7)$$

This is justified for instance by the computer simulations presented by Kiritani et al. demonstrating that even at the lowest end of the temperature range in which HVEM loop-growth experiments are carried out in practice the time it takes to build up steady-state conditions is short compared with the duration of an experiment. In the experiments on loop growth in copper to be quoted in the present work the lowest irradiation temperature was 160°C for which the build-up time is less than 10 seconds.

Foreman calculated the vacancy concentration in the interior of an infinitely thick foil to be

$$C_V^{\infty} = (P \gamma_I / (Z_{IV} v_V \gamma_V))^{\frac{1}{2}} \quad (8)$$

Inserting the Foreman factor $g(f,d)$ and equations (5), (6), (7),

and (8) in equation (1) under the assumption that the mobilities M_I and M_V in (1) are identical to the jump frequencies ν_I and ν_V (the exact meaning of the term mobility was not specified by Kiritani et al.) produces our final equation for the loop growth rate:

$$\dot{L} = 2\sigma(z_{IL}\gamma_V/\gamma_I - z_{VL}) \cdot g(f,d) \cdot (P\gamma_I\nu_V/(z_{IV}\gamma_V))^{\frac{1}{2}} \quad (9)$$

For the interior of an infinitely thick foil, i.e. for $g(f,d)=1$, equation (9) is very close to equation (2); if $\gamma_I=\gamma_V=1$ as assumed by Foreman, the two equations are identical. Thus, for the interior of an infinitely thick foil equation (9) gives the same loop growth rate as that quoted by Kiritani et al. [1] for the vacancy-dominant case.

For the other extreme situation (small values of $g(f,d)$ in equation (9) and the surface-dominant case of Kiritani et al. as described in equation (3)) a similar simple analytical comparison is not possible: Foreman does not give any analytical expression for $g(f,d)$, and Kiritani et al. do not define C_S in equation (3) in terms of the fundamental parameters. However, a comparison of the \dot{L} values obtained from equation (9) for f equal to 1 and f equal to 10, the variation in f being produced by variation in ν_V or P , gave almost the same types of dependence on ν_V and P as those expressed in equation (3): no dependence on ν_V and proportionality with P .

4. The Relation between Apparent Activation

Energy for Loop Growth and E_V^M

On the basis of fig. 1 and equation (9) it is possible to construct a generalized chart for the relation between the

apparent activation energy for loop growth (from now on designated E) and the vacancy migration energy E_V^M . In an experiment to determine E all parameters but the temperature will be constant. A sufficiently large change in irradiation temperature will, via the change in v_V in equation (4), make f vary from the low values typical of the surface-dominant case to the values of 10^5 or more typical of the vacancy-dominant case ($g(f,d) \sim 1$).

Let us consider an experiment in which f takes the values 1, 10, 10^2 , 10^3 , 10^4 , 10^5 , and 10^6 , i.e. the values for which Foreman has calculated $g(f,d)$ as shown in fig. 1. We can rewrite equation (4) as

$$f(n) = k_1 e^{E_V^M/kT(n)} = 10^n \quad (10)$$

with n taking the values 0, 1, 2, 3, 4, 5, and 6 and k_1 being a constant. The fulfilment of equation (10) requires that

$$E_V^M/kT(n) = n \cdot \ln 10 - \ln k_1 = n \cdot \ln 10 + E_V^M/kT(0) \quad (11)$$

For our purpose it is irrelevant by which combination of the parameters k_1 and E_V^M these equations are fulfilled.

In order to determine E we would plot $\ln \dot{L}$ versus $1/kT$. E is then the absolute value of the slope. By combining equations (9), (10), and (11) we get the following expression for $\ln \dot{L}(n)$ in our experiment (k_2 being a constant):

$$\ln \dot{L}(n) = k_2 + \ln(g(10^n, d)) - n/2 \cdot \ln 10 \quad (12)$$

Thus, if we plot $\ln \dot{L}(n)$ from equation (12) versus n , we are actually doing an activation-energy plot with one unit along the n axis corresponding to $\ln 10/E_V^M$ units of $1/kT$ (according to equation (11)). Such plots are shown in fig. 2 for positions 1/2, 1/4, and 1/10 foil thickness away from the surface (i.e. for d values of 0.5, 0.25, and 0.1). The plotted points are connected with straight lines; the absolute values of the slopes of these lines

are given in table 1. It should be underlined that fig. 2 does not represent a basically new approach to loop growth. The basic model is still that of Kiritani et al. [1] - but now with the theoretically indisputable improvement that the point-defect profile in the thin foil is included in a systematic way.

The significance of fig. 2 is that the transition range between the surface-dominant case with $E=0$ and the vacancy-dominant case with $E=1/2 E_V^H$ covers a very wide range of f values - and that, as it is to be shown in section 5, these values correspond to typical experimental situations. Furthermore, the deviation from linearity is so small in the upper end of the transition range that it would not be detected in the typical experiment, because the range covered is too narrow; the plot of $\ln L$ versus $1/kT$ would look like a straight line with a slope lower than $1/2 E_V^H$. Already Urban and Wilkens [7] referred to the possibility of a transition range with E values below $1/2 E_V^H$ (they quoted an E range of $1/4 E_V^H - 1/2 E_V^H$). However, they did not specify any relation between E and the other physical parameters.

It should be noticed that fig. 2 is based on values of $g(f,d)$ taken from fig. 1. This procedure will introduce some error particularly for low f values and the d values 0.25 and 1.1. The exact slopes of the lines in fig. 2 as given in table 1 may thus be questionable, but this is of no practical importance for our use of fig. 2 and table 1.

5. Comparison with Experimental Results and Discussion

As already mentioned Kiritani et al. [1] quoted an E value (apparent activation energy for loop growth) of 0.30eV for copper.

They also showed activation-energy plots for various foil thicknesses without quoting the slopes. The same plots were shown by Kiritani [8], but this time with slopes quoted. These plots are reproduced in fig. 3. There is a trend for E to increase with increasing foil thickness (or increasing f , cf. table 2), particularly when one omits the $0.18\mu\text{m}$ foil for which the slope is based on very few points. Fig. 3 indicates that the E value of 0.30eV quoted by Kiritani et al. refers to quite thin foils.

Recently Barlow, Leffers, and Singh [9] found an E value of 0.35eV in comparatively thick foils of Cu (and dilute Cu-Ni alloys). This result as well as the results in fig. 3 can be ascribed to an activation energy for vacancy migration (E_V^M) of $\sim 0.7\text{eV}$ with corresponding E values that vary with f according to fig. 2. To check this we have in table 2 listed the E values for Cu from fig. 3 and from [9] divided by E_V^M together with the experimental parameters and the derived values of Foreman's f parameter. The uncertainties quoted for the results from fig. 3 are derived from the E uncertainty of $\pm 0.02\text{eV}$ quoted by Kiritani et al; in [9] the standard deviation on E was also found to be about 0.02eV . In the calculation of f we have used $z_{IV}=10$ [6], $v_V=10^{13} e^{-E_V^M/kT_s} \text{s}^{-1}$ [1], $E_V^M=0.70\text{eV}$, $\gamma_I=\gamma_V=1$ [6], $\lambda=0.255\text{nm}$.

In table 2 E/E_V^M is significantly smaller than 0.5 for $f < 2 \cdot 10^4$. This is in perfect agreement with fig. 2/table 1: if we consider loops in the centre of the foil ($d=0.5$), E/E_V^M becomes significantly smaller than 0.5 in the f range 10^3 - 10^4 ; if we consider loops halfway between the foil centre and the surface ($d=0.25$), the deviation from $E/E_V^M=0.5$ becomes significant in the f range 10^4 - 10^5 .

This comparison of table 2 with fig. 2/table 1 illustrates the use and the limitations of fig. 2. Qualitatively fig. 2

satisfactorily explains the observation of E/E_V^M values below 0.5. On the other hand one cannot in practice get an exact value for E/E_V^M from fig. 2. With reference to fig. 3 the problem would arise already because we do not know the exact value of d . However, even if d is known and the curves in fig. 2 are made smooth by the addition of points for non-integer values of f , there are other difficulties: i) the exact value of Z_{IV} and of the preexponential factor in v_V are not known; ii) there is some ambiguity about the values of γ (Johnson and Lam [10] for instance used $\gamma_I=2/3$ instead of 1); iii) there is also some ambiguity about the calculation of the dose rate P from the beam current (for instance Kiritani et al. [1] refer to the problem of close-pair recombination). In practice fig. 2 can therefore be used to check whether the experimental conditions are such that E/E_V^M is sufficiently close to 0.5, but it cannot be used to calculate E_V^M from E when E/E_V^M is significantly smaller than 0.5. However, these limitations in the use of fig. 2 does not change the basic fact that fig. 2 produces a convincing explanation of the observation of E/E_V^M values smaller than 0.5.

For certain experimental situations the loop growth rate becomes practically independent of temperature ($E=0$) [1,2,8]; this is the pure surface-dominant case (cf. section 2). According to fig. 2 this situation should be reached for $f \approx 1-10$. In table 3 we have listed the experimental parameters and the f values for the actual transition points for the Cu results from fig. 3 (f is calculated with the same values of the constants as used in table 2).

The experimental results in table 3 and fig. 3 clearly differ from the theoretically predicted behaviour in fig. 2: the f value for which loop growth becomes temperature-independent is 10^2-10^3

times bigger than it should be according to fig. 2, and there is a much sharper transition between the temperature-dependent and the temperature-independent region than indicated in fig. 2.

As already mentioned there are some uncertainties involved in the calculation of f . However, these uncertainties cannot lead to errors of 2-3 orders of magnitude in the calculated f values. Furthermore, the agreement between experimental and calculated values of E/E_V^M for $f \geq 3 \cdot 10^3$ indicates that the calculated f values are reasonably correct. Thus the discrepancies between table 3/fig. 3 and the theoretical predictions in fig. 2 cannot be explained simply in terms of errors in the calculated f values.

Kiritani et al. [1] quote an approximate expression for the "critical thickness", which is the thickness where the transition from temperature-dependent to temperature-independent loop growth should take place in a given material at a given temperature and dose rate, and they find a satisfactory agreement with their Cu experiments. However, what they present is an approximate solution to the set of equations describing the physical situation common to their work and the present work, whereas we, in fig. 2, present the exact solution to these equations. Thus, when they find a satisfactory agreement with their experimental results and we do not, this simply illustrates the limitation in their approach. Their critical thickness refers to the situation where the loss of point-defects to the surface is equal to the recombination loss, i.e. it should correspond to the middle of the theoretical transition range in fig. 2, which it actually also does.

Logically, these discrepancies can have their origin either in limitations in the correctness of Foreman's calculated profiles in fig. 1 or in limitations in the correctness of equation (1). There is no obvious source of error in Foreman's calculations

(as to the possibility that there is a significant effect of divacancies see section 6), whereas there are two basic shortcomings in equation (1): it does not consider the long-range interaction between the point defects and the dislocation, and it does not consider the local distortion of the point-defect population around the dislocation. Therefore, the most likely explanation of the discrepancies is that equation (1), even though it applies approximately for $g(f,d)$ values close to 1, does not give a correct description of loop growth when $g(f,d)$ is closer to zero (small effective distance to the surface).

6. The Possible Effect of Divacancies

So far we have neglected the possible effect of divacancies. Kiritani et al. [1] suggested that values of E/E_V^M smaller than 0.5, as they observed in copper, are due to divacancies with higher mobility than monovacancies. As shown in the present work, these observations can be accounted for by the point-defect profile in the thin foil, which removes the necessity of involving divacancies. But, of course, this does not remove the possibility that divacancies can play a role in the loop-growth process.

In order to evaluate the effect of divacancies on loop growth one needs to consider two parameters: the concentration of divacancies and their mobility. It is obvious that the effect of divacancies would become significant only if a sufficiently large population of (fast moving) divacancies exists during the loop-growth experiments; if the divacancies dissociate to re-form monovacancies under the relevant experimental conditions, their influence can be neglected.

In the case of copper there is no clear and direct experimental evidence available to give reliable values of the binding and migration energy for divacancies. In a recent review Balluffi [11] for instance has abstained from quoting any divacancy binding and migration energy for Cu. From stage III recovery experiments Antesberger, Sonnenberg, Wienhold, Coltman, Klabunde, Williams, and Chaplin [12] concluded that divacancies in Cu are not significantly more mobile than monovacancies. Recently Wienhold, Sonnenberg, and Antesberger [13] have reanalyzed basically the same experimental results as quoted by Antesberger et al. They claim that the results can be consistently interpreted on the basis of either of the following two assumptions: i) divacancies are much more mobile than single vacancies and are tightly bound (i.e. they do not under the given experimental conditions dissociate to any significant extent), or ii) the migration energies of single- and divacancies are very similar.

Kiritani et al. [1] have included divacancies in their rate equations for loop growth, assuming that the conditions correspond to case i) of Wienhold et al. Under these conditions Kiritani et al. found the apparent activation energy for loop growth E to be

$$E = (E_V^M + E_{2V}^M) / 4 \quad (13)$$

E_{2V}^M being the divacancy migration energy. Equation (13) refers to conditions away from the surface in a sufficiently thick foil. If we take case i) of Wienhold et al. to mean that E_{2V}^M for Cu should be 0.50eV, the resulting E value is ~ 0.30 eV. This does not agree with the observations quoted in table 2 of E values of ~ 0.35 eV in Cu foils of sufficient thickness (E is E/E_V^M in table 2 multiplied by 0.70eV) - particularly not in view of the fact that the obvious experimental errors (the foils being thinner than estimated, the loops being too close to the foil surface) can only bring the

measured E values down.

The absence of observable divacancy effects in the loop-growth experiments on Cu can have two possible explanations: either divacancies in Cu do not have significantly lower migration energy than monovacancies, or the concentration of divacancies is very low during the HVEM loop-growth experiments. The former is an obvious explanation that cannot be ruled out on the basis of existing evidence (cf. Wienhold et al.). In order to evaluate the latter we shall have to take a closer look at the rate equations that Kiritani et al. [1] derive for the case with a strong effect of divacancies. They find the following expression for the concentration of divacancies C_{2V} :

$$C_{2V} \approx 1/\sqrt{2} (Z_{VV}/Z_{I2V})^{1/2} (M_V/M_{2V})^{1/2} C_V \approx (M_V/M_{2V})^{1/2} C_V \quad (14)$$

where the last approximation is made by the present authors. In this equation Z_{VV} and Z_{I2V} are the capture-site number for the reaction between two vacancies to form a divacancy and the reaction between an interstitial and a divacancy to form a monovacancy, respectively, and M_{2V} is the divacancy mobility. A combination of the equations (8) and (11) of Kiritani et al. and our equation (14) with the assumption $M_I \gg M_{2V} \gg M_V$ produces an expression for the monovacancy concentration

$$C_V \approx (P/Z_{IV})^{1/2} (M_V M_{2V})^{-1/4} \quad (15)$$

As already mentioned, Kiritani et al. claim to consider a situation where the dissociation of divacancies to monovacancies is insignificant. Whether this is actually the case can be decided on the basis of equations (14) and (15) and Damask and Dienes's equation for the equilibrium between monovacancies and divacancies [14]

$$C_{2V}/C_V^2 = 6e^{E_{2V}^B/kT} \quad (16)$$

E_{2V}^B being the divacancy binding energy (and the vibrational entropy term being neglected). If the ratio C_{2V}/C_V^2 derived from equations (14) and (15) is clearly smaller than $6e^{E_{2V}^B/kT}$, i.e. if there is a surplus of monovacancies relative to divacancies, Kiritani et al. are justified in neglecting divacancy dissociation: such a situation would mean that the divacancies are removed by reaction with other defects before they dissociate to any significant extent, because the monovacancies cannot combine sufficiently fast to produce equilibrium concentration of divacancies. If the ratio is not clearly smaller than $6e^{E_{2V}^B/kT}$, the equations of Kiritani et al. are not selfconsistent, exactly because they have neglected divacancy dissociation. Hence, the equations of Kiritani et al. would be consistent only if

$$(P/Z_{IV})^{-1/2} M_V^{3/4} M_{2V}^{-1/4} \ll 6e^{E_{2V}^B/kT} \quad (17)$$

or, expressed with a ratio R, if

$$R = (P/Z_{IV})^{-1/2} M_V^{3/4} M_{2V}^{-1/4} / (6e^{E_{2V}^B/kT}) \ll 1 \quad (18)$$

In the case of Cu we have already used numerical values for most of the factors in condition (17) or (18). In the following we shall use $Z_{IV}=10$, $M_V=10^{13}s^{-1}e^{-0.70eV/kT}$, $M_{2V}=10^{13}s^{-1}e^{-0.50eV/kT}$, $E_{2V}^B=0.20eV$. The E_{2V}^B value of 0.20eV is estimated on the basis of the E_{2V}^B values quoted for other f.c.c. metals in Balluffi's review (with $E_{2V}^B=0.20eV$, the value of E_{2V}^B/E_V^M for Cu becomes equal to the average of the E_{2V}^B/E_V^M values for the other f.c.c. metals for which E_{2V}^B is quoted). In the Cu loop-growth experiments of Kiritani et al. the mean temperature was 200°C and the dose rate $1.6 \cdot 10^{-3}s^{-1}$. When these values are inserted, R in condition (18) gets a value of ~17, i.e. the condition is very far from being fulfilled. This means that the divacancy concentration derived from the rate equations of Kiritani et al. is far too high compared with the

monovacancy concentration and hence that the assumptions on which equation (13) is based are not valid.

The absence of any observable effect of divacancies in the Cu loop-growth experiments is therefore not surprising: even if E_{2V}^M is lower than E_V^M in Cu, one would not expect that equation (13) is obeyed, because the divacancies, contrary to the assumption of Kiritani et al., largely disappear by dissociation under the given experimental conditions.

Up to this point we have only considered the possible divacancy effect on the growth of loops away from the surface in sufficiently thick foils (in our terminology for $g(f,d)=1$). However, when divacancies do not have any significant effect in Cu under such circumstances, there is no reason to believe that they should have a significant effect for $g(f,d)<1$: with decreasing $g(f,d)$ the point-defect contrations decrease, which will change the equilibrium monovacancies/divacancies even further towards monovacancies (cf. equation (16)).

In order to evaluate the applicability of our approach to loop growth in the EVEM, in which we neglect the effect of divacancies, to other metals (with $E_{2V}^M < E_V^M$) one has to apply condition (17) or (18) to the metal in question. If they are fulfilled, it is not justified to neglect divacancies; if they are far from being fulfilled, i.e. if for instance R in condition (18) is much bigger than 1, our approach as summarized in fig. 2 should apply. In table 4 we have calculated R for the metals for which Balluffi [11] has quoted E_V^M , E_{2V}^M , and E_{2V}^B . In the cases where a range is quoted we have taken the midpoint of the range except for E_{2V}^B in gold where we have done the calculations for the two extreme values of the range quoted. The calculations have been done for $T/T_M=0.35$ (T_M being the melting point) corresponding to 200°C for Cu which is the average

temperature in the experiments of Kiritani et al. [1] and for $T/T_M = 0.42$ corresponding to 300°C for copper which is about the average temperature in the experiments of Barlow et al. [9]. We have taken P to be $1.6 \cdot 10^{-3} \text{ s}^{-1}$, Z_{IV} to be 10, and the preexponential factor in the M 's to be 10^{13} s^{-1} . The results for copper are also given in table 4.

Table 4 demonstrates that there is no general answer to the question whether the divacancies can be neglected in HVEM loop-growth experiments as we have done in our generalized chart in fig. 2. If the parameters involved are known with sufficient accuracy, one can in each individual case get a theoretical answer from condition (17) or (19) (with inverted signs of inequality when used as conditions for the applicability of fig. 2). Experimentally, the situations where the divacancies can be neglected are characterized by an apparent activation energy for loop growth of $1/2 E_V^M$ for loops away from the surface in sufficiently thick foils. Table 4 indicates that the situation where one gets the full effect of divacancies as expressed in equation (13), characterized by R values much smaller than 1, is the exception rather than the rule.

In the cases where a significant divacancy effect is to be expected ($M_{2V} \gg M_V$, $R \ll 1$), there is a high probability that the divacancies react with monovacancies to form trivacancies. If the trivacancies have migration and binding energies similar to those of divacancies, they will in turn react to form tetravacancies. This chain reaction will continue until the clusters reach a size where they become immobile and hence lose their importance for vacancy transport (which probably happens at the tetravacancy stage). Thus, equation (13) will probably overestimate the effect of divacancies even for $M_{2V} \gg M_V$ and $R \ll 1$, because the population of mobile vacancy clusters will be eroded by the above chain reaction.

7. Conclusions

It is shown theoretically that there is, within the basic framework of the work of Kiritani et al. [1], a relatively wide transition range between the vacancy-dominant and the surface-dominant case.

It is demonstrated that the transition range corresponds to ordinary experimental situations and that this can account for the observation of apparent activation energies for loop growth smaller than half the vacancy migration energy.

In the "lower" end of the transition range, where point-defect loss to the surface becomes comparable to the recombination loss, the exact agreement between experiment and theory ceases: in the experiments there is a sudden transition to a behaviour typical of the surface-dominant case (temperature-independent loop growth) rather than the continued smooth transition predicted by theory.

The conclusions above are based on calculations in which the effect of divacancies have been neglected. All comparisons with experiments have been made for copper, and the theoretical and experimental evidence is that it is justified to neglect the divacancies in this metal. We suggest general theoretical and experimental criteria to decide whether or not it is justified to neglect the divacancies in a specific experiment with a specific material.

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Table 1

Absolute values of the slopes of the straight sections in fig. 2 (in units of E_V^*)

logf interval	d=0.1	d=0.25	d=0.5
0-1	0.0001	0.0259	0.0421
1-2	0.1332	0.1615	0.1754
2-3	0.2747	0.3403	0.3886
3-4	0.3319	0.4349	0.4771
4-5	0.4031	0.4836	0.4944
5-6	0.4628	0.4967	0.5000

Table 2

E (apparent activation energy for loop growth) from various experiments on Cu divided by E_V^M (0.70eV) and the corresponding experimental parameters and values of the Foreman parameter f

source	E/E_V^M	foil thickness (μm)	mean temperature for the sloping part of the plot ($^{\circ}\text{C}$)	dose rate (dpa/s)	f
[8]	0.43 ± 0.03	0.12	190	$1.6 \cdot 10^{-3}$	$3.2 \cdot 10^3$
[8]	0.43 ± 0.03	0.16	200	$1.6 \cdot 10^{-3}$	$7.0 \cdot 10^3$
[8]	(0.39*)	0.18	220	$1.6 \cdot 10^{-3}$	$5.6 \cdot 10^3$
[8]	0.44 ± 0.03	0.22	220	$1.6 \cdot 10^{-3}$	$1.2 \cdot 10^4$
[8]	0.51 ± 0.03	0.25	210	$1.6 \cdot 10^{-3}$	$2.9 \cdot 10^4$
[8]	0.47 ± 0.03	0.35	210	$1.6 \cdot 10^{-3}$	$1.1 \cdot 10^5$
[9]	0.50 ± 0.03	~ 0.5	325	$1.3 \cdot 10^{-2}$	$1.5 \cdot 10^5$

* based on 2-3 points only

Table 3

Experimental parameters and f values for the transition point from temperature-dependent to temperature-independent loop growth (fig. 3)

foil thickness (μm)	temperature ($^{\circ}\text{C}$)	f
0.12	220	$1.0 \cdot 10^3$
0.16	240	$1.8 \cdot 10^3$
0.18	245	$2.5 \cdot 10^3$

Table 4

Calculated values of R from condition (18) for the metals for which Balluffi [11] has quoted E_{V}^{M} , $E_{2\text{V}}^{\text{M}}$, and $E_{2\text{V}}^{\text{B}}$ and for Cu

Metal	$T/T_{\text{M}}=0.35$	$T/T_{\text{M}}=0.42$
Au ($E_{2\text{V}}^{\text{B}}=0.25\text{eV}$)	0.7	16
Au ($E_{2\text{V}}^{\text{B}}=0.57\text{eV}$)	$3 \cdot 10^{-4}$	0.02
Al	0.08	3
Pt	6	95
Cu	17	223

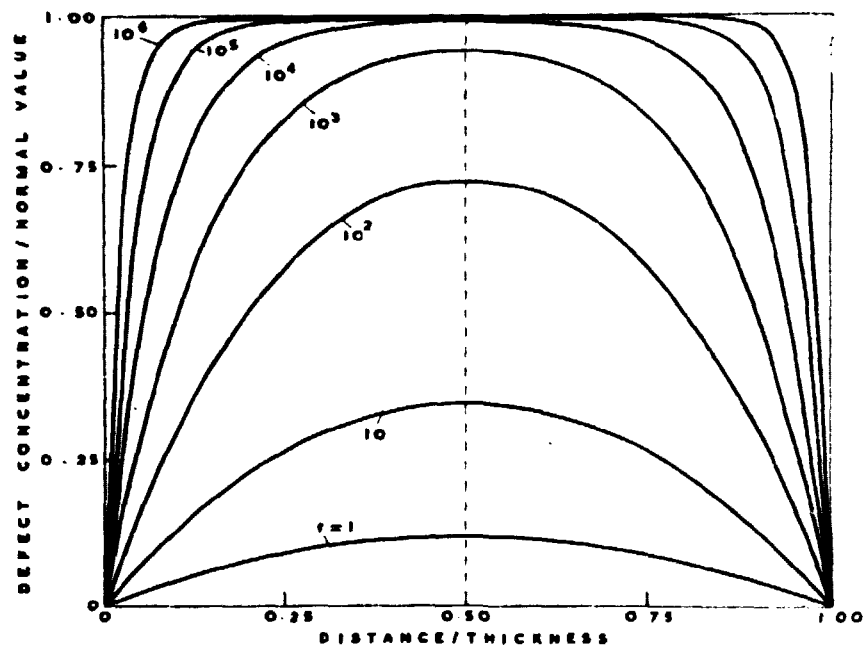


Fig. 1. The Foreman proportionality factor $g(f,d)$ as a function of f (equation (4)) and d (distance from foil surface divided by foil thickness). The point-defect concentration for a given combination of f and d is equal to the concentration in the central part of an infinitely thick foil multiplied by $g(f,d)$. From [6].

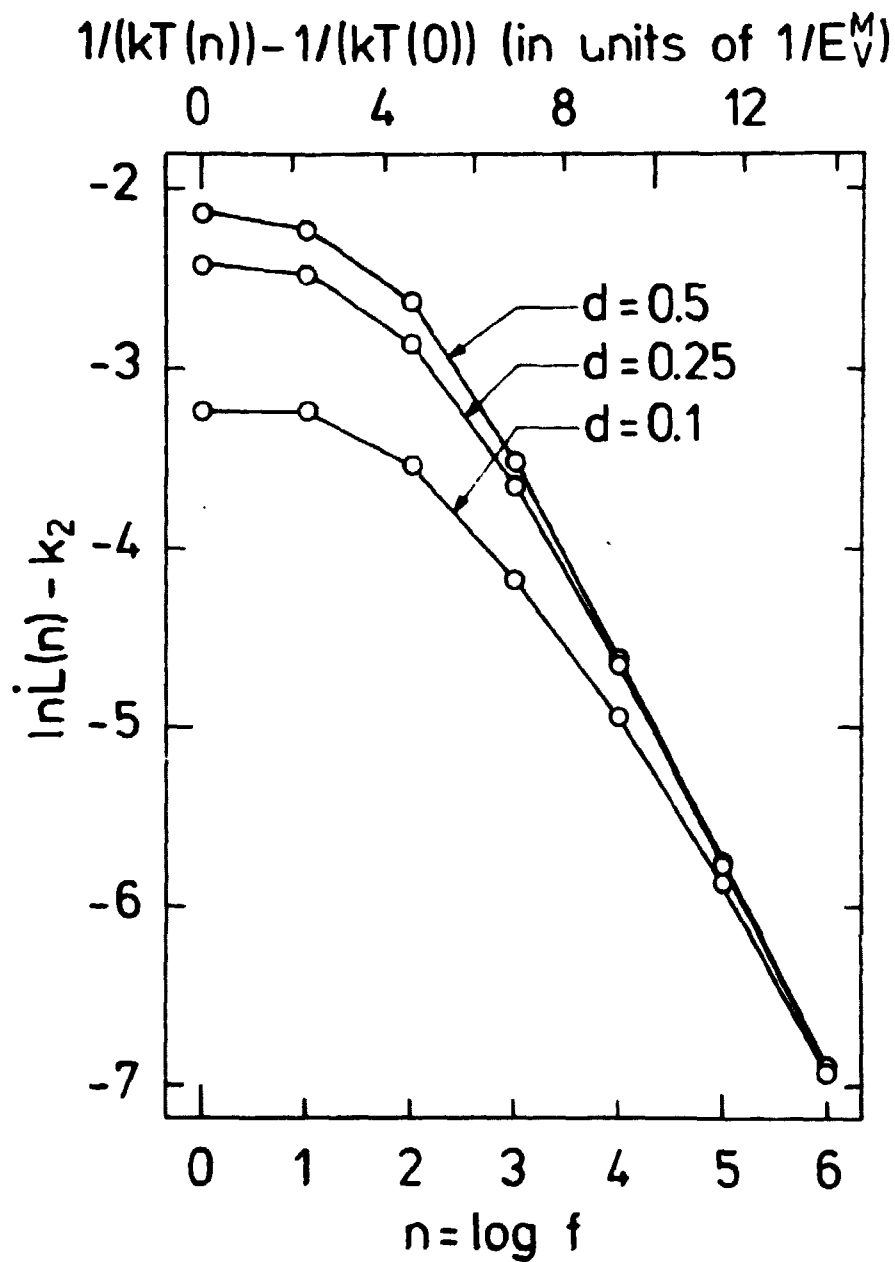


Fig. 2. Generalized theoretical activation-energy plot for three different positions in the foil. Absolute slopes for the individual straight sections are given in table 1.

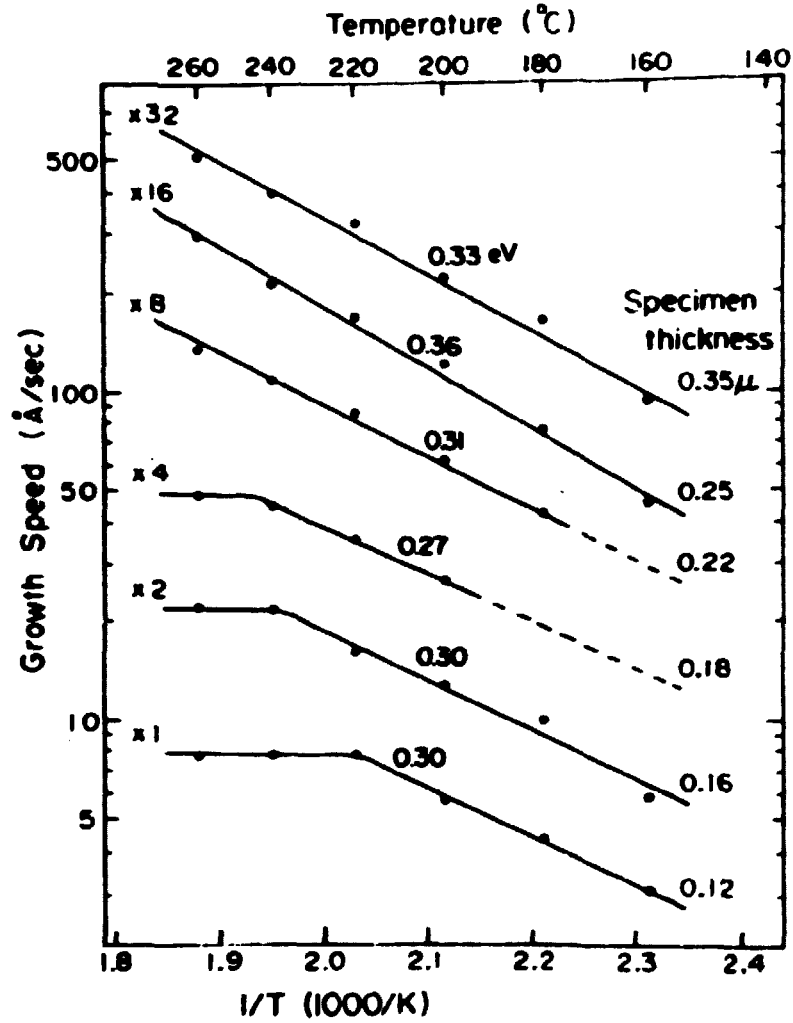


Fig. 3. Logarithmic loop growth rates in Cu foils of different thicknesses versus $1/T$. The slope (apparent activation energy) is given for each foil thickness. In order to separate the individual experiments different scaling factors (1, 2, 4, 8, 16, 32) have been used for each experiment. From [8].